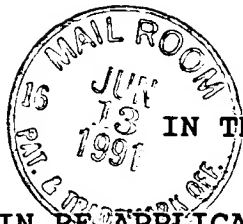


017714441



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION TO REISSUE
U.S. PAT. NO. 4,912,155,
ISSUED MARCH 27, 1990

SERIAL NO.

FILED JUNE _____, 1991

FOR ANTIOXIDANT AROMATIC FLUORO-
PHOSPHITES

Express Mail mailing label
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Date of Deposit JUNE 13, 1991
I hereby certify that this paper or fee is being
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is addressed to the Commissioner of Patents and
Trademarks, Washington, D.C. 20231
DEBORAH REAGAN
(Typed or printed name of person mailing paper or
fee)
Deborah Reagan
(Signature of person mailing paper or fee)

**REISSUE PETITION, DECLARATION
AND POWER OF ATTORNEY**

I, Lester P.J. Burton, residing in New Castle County, State of Delaware, and citizen of Canada, hereby declare that:

1. I believe I am the original, first and sole inventor of the invention entitled ANTIOXIDANT AROMATIC FLUOROPHOSPHITES, described and claimed in the reissue application which is attached hereto, and which was originally filed on February 27, 1987 as Application Serial No. 20,023, and which issued as U.S. Pat. No. 4,912,155 on March 27, 1990.

2. I have read and understand the contents of the above-identified specification, including the claims.

3. I hereby request that I may be allowed to surrender and do hereby assent to surrender the said U.S. Pat. No. 4,912,155, which is assigned in whole to my former employer, Ethyl Corporation, and request that the Patent may be reissued, upon the foregoing claims.

4. I believe that the Patent may be wholly or partly

inoperative or invalid by reason of my claiming more or less than I had a right to claim in the Patent; and that this was the result of error without any deceptive intention on the part of myself or my employer and assignee of the Patent. The errors relied upon are set forth in the paragraphs below, together with a specification of how and when the errors occurred.

5. I was unaware of any errors in the Patent until April 2, 1991, when I met with counsel, and certain errors specified below were identified.

Claim 1

6. I was assigned the task by my employer to discover new and useful antioxidants for organic materials, mainly polyolefin polymers, especially polypropylene. In the course of my work, I discovered aromatic fluorophosphites that were surprisingly good antioxidants and most surprisingly had far better hydrolytic stability than commercial antioxidants.

7. During the development of my invention, I had a librarian conduct a literature search to determine the state of the art. The search uncovered no indications that any aromatic fluorophosphites had ever been disclosed as antioxidants. Several aromatic fluorophosphite chemical structures had been disclosed in the prior art, but none of these were indicated to have any utility to the best of my knowledge. I summarized my results in a memo that on October 8, 1986, I sent to Joseph Odenweller, the attorney then responsible for prosecuting my patent application. That memo is attached as Attachment A.

8. I carefully reviewed the specifications and claims of the patent application which Mr. Odenweller prepared. However, in reviewing the claims, I failed to compare the claims against the prior art structures that I had previously uncovered. Having disclosed the structures to the attorney handling the patent application, I simply assumed that the attorney had taken the necessary steps in view of my invention and the prior art. I have limited knowledge of patent law, and relied on the attorney handling the prosecution to make certain that all patent law requirements were met.

9. I am now informed that Mr. Odenweller had apparently misunderstood my memo Attachment A. The memo summarized the results of an STN International computerized search. The results provide the structure, and, if the structure appeared only before 1967 there are "0 references" cited, but if the structured appeared in 1967 or after, references are cited. In my memo Attachment A, I had indicated for some structures that there were "0 ref", which meant as indicated above. I am informed that Mr. Odenweller understood that "0 ref" meant that the structure did not appear in the prior art. Unfortunately, I never discussed this memo with any attorney during the prosecution, and, therefore, never explained the import of my notations. Upon further review of the literature search results, I have also discovered that I overlooked and omitted in my memo certain aromatic fluorophosphite chemical structures that were in the computerized survey. I am now also informed that the attorney inadvertently failed to take into

account all the prior art chemical structures that I did supply to him. I believe that these inadvertent failures led to claim 1 of the Patent to potentially claim chemical compounds that were disclosed in the prior art.

10. I have now also been shown certain prior art of which I have no recollection of being aware at the time that my original patent application was filed. That art is stated in the Information Disclosure Statement that I understand is being filed together with this petition.

11. More specifically, I verily believe that claim 1 Formulas III and IV may claim chemical compounds which were in the above prior art disclosures, and should be further limited.

12. Further, with respect to Claim 1, it has now been brought to my attention that the aromatic substituents in claim 1 Formula V are limited to tert-alkyl groups, and in Formula VI are limited to certain substituents. In the specifications, I had disclosed that the substituents in both formulas could be groups other than the tert-alkyl groups specified for Formula V or the groups which were specified for Formula VI. My invention is broader than that shown in these formulas, and I believe that the patent may be partly inoperative in specifying my invention by the inadvertent failure to include the other substituents. I do not recall there being any reason for the limitation to the groups presently specified in the claims, and, for reasons described before, I did not focus on the discrepancy between the specification and this claim. The amendments are believed to include the substituents

described in the disclosure, without resulting in any compounds described in the prior art.

Claim 6

13. The present Claim 6 utilizes nomenclature which is accurate, but is not the best way of describing the compound. The language has been amended to utilize more descriptive nomenclature.

Claim 8

14. Claim 8 is being amended in view of U.S.S.R. Authorship Certificate 398,574. The amendment is to make more clear that the antioxidants of the invention are added to the organic materials by mixing or spraying and are not substantially reacted with the organic materials, as is shown in the U.S.S.R. Authorship Certificate. I first became aware of the U.S.S.R. Authorship Certificate on April 2, 1991. I now recognize that the use of the term "containing" may potentially cause the claim to read on the reaction of antioxidant with the organic composition, which was not my intent.

Claim 23 and 24

15. Typographical errors in claims 23 and 24 are corrected. The errors were not my fault, but arose in the printing of the original patent by the Patent and Trademark Office.

Claim 26

16. In Claim 26, Structure IV (vi), and "0" is missing in the formula, and should be corrected to conform to the specification (see the original patent, Col. 11, lines 30-40).

17. The specification contains corrections to typographical

errors in the printed original patent, which errors were not my fault but arose in the printing by the Patent and Trademark Office.

18. At Cols. 15 and 16, I have also deleted the disclosure of the alternate use of PBr_3 in making an intermediate in the making of my invention. At the time I originally filed my application I believed that PBr_3 might be a useful reactant. I have since discovered that PBr_3 does not appear to work in the reaction.

19. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

20. I hereby appoint the following attorneys or agents to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith: Rudolf E. Hutz, Reg. No. ³⁰¹22,397; Thomas M. Meshbesher, Reg. No. 30,982; Robert G. McMorrow, Reg. No. 30,962; Philip M. Pippenger, Reg. No. 25,525 and Richard L. Hansen, Reg. No. 27,338.

Address all telephone calls to Rudolf E. Hutz at telephone no. (302) 658-9141.

Address all correspondence to:

601 Rudolf E. Hutz
602 Connolly, Bove, Lodge & Hutz
701 1220 Market Building
702 P.O. Box 2207
703 Wilmington, Delaware 19899

Date: June 10, 1991


Lester P.J. Burton 40100

Country of Citizenship: Canada
Residence: 2703 Tanager Road,
Wilmington, Delaware DE

300,

10/8/86

The following represents
what I know & to please
with. If you want to
see the actual record, let
me know.

LPTB

Attachment A
to
Declaration of Lester P.J. Burton

ETHYL CORPORATION

INTER-OFFICE

To K A K ebllys

ADDRESS ETC

FROM LPT Burton

ADDRESS ETC

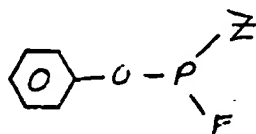
SUBJECT Fluorophosphites

DATE 12/10/85

A computer search of the substructure PhOPF was executed. The search yielded 171 structures and 52 references. The majority of the hits were metal complexes, phosphates or pentacoordinate species.

No reference to antioxidant activity was found.

The arylfluorophosphites found are listed below:



a) $\text{Z} = \text{O}-\text{C}_6\text{H}_5$ 3 ref

b) $\text{O}-\text{C}_6\text{H}_4$ 0 ref

c) $\text{O}-\text{C}_6\text{H}_3$ 1 ref

d) $\text{O}-\text{C}_6\text{H}_4$ 0 ref

e) OMe 0 ref

f) $\text{O}-\text{Pr}$ 0 ref

g) $\text{O}-n\text{-Bu}$ 0 ref

Ar_2PF

h) $\text{Ar} = \text{O}-\text{C}_6\text{H}_4$ 0 ref

i) $\text{O}-\text{C}_6\text{H}_4$ 0 ref

(RR'N)₃P (NHR' could be in place of RR'N). Named in connection with the antistatic treatment of plastics.¹⁴⁴
 s-Hexamethylene-diaminophosphite. Named in connection with the production of profiled plastic strips which are weather and light stable.¹¹⁹

O,O-diethyl N,N-diethylphosphoroamidite. Used for the preparation of pesticides.⁴⁸

Amides and imides of phosphorous acid. Catalyze the polymerization of formaldehyde to high molecular weight poly(oxy methylenes).¹²⁷

Polycondensates from PCl₃ and such as hexamethylene diamine. Applicable as ion exchangers.²⁴⁸

(R₂N)₃P, R = Me, Et, Pr, Bu, especially Me. Improve leaded gasoline with regard to preignition and octane number.⁶²

Di-Et N-(2,4-diMe-phenyl) phosphoramidite. Is an additive for motor gasoline, improving the octane number, and minimizing combustion zone deposits without lowering the octane number.²⁴¹

"N,N',N''-(trioctylphenyl)phosphorous triamide" "Di-Bu-N-phenyl amidophosphite." Named as antiknock additives for gasoline.²⁸⁹

PhOP(NCO)₂. Named in connection with pigmented polyurethane coating compositions having improved viscosity stability.⁶²⁰

I. LIST OF COMPOUNDS

I.1. Phosphites

I.1.1. Difluorophosphites

TYPE: ROPF₂

CH₃OPF₂. ³¹P -111 ppm, J_{PF} 1275 Hz.⁶²⁸

F₂POCH₂CH₂OPF₂. (CH₂OP)₂Cl₄ + SbF₃. b₁₈₀ 50°, n_D²⁶ 1.3523, ¹⁹F NMR, ¹²⁴⁹ ³¹P -112.0 ppm, J_{PF} 1295 Hz.¹²⁰⁹, ¹²⁴⁹

PROPF₂. ROPCl₂ + SbF₃. b. 44.5°, n_D²⁰ 1.3400, ¹⁹F NMR, ¹²⁴⁹ ³¹P -111.5 ppm, J_{PF} 1287 Hz.¹²⁰⁹, ¹²⁴⁹

CH₂:CHCH₂OPF₂. ROPCl₂ + SbF₃. b. 42°, ¹⁹F NMR, ¹²⁴⁹ ³¹P -111.9 ppm, J_{PF} 1290 Hz.¹²⁰⁹, ¹²⁴⁹

BuOPF₂. ROPCl₂ + SbF₃. b. 75°, n_D²⁰ 1.3580, ¹²⁴⁹ ³¹P -111.9 ppm, J_{PF} 1288 Hz.¹²⁰⁹

PhOPF₂. PhOPCl₂ + SbF₃. b₆₀ 58°, n_D²⁷ 1.4575, ¹⁹F NMR, ¹²⁴⁹ ³¹P -110.1 ppm, J_{PF} 1326 Hz.¹²⁰⁹, ¹²⁴⁹

1,4-C₆H₄(OPF₂)₂. -PCl₂ + SbF₃. b₁₂ 59°, n_D²³ 1.4488, ¹⁹F NMR, ¹²⁴⁹ ³¹P -109.8 ppm, J_{PF} 1328 Hz.¹²⁰⁹, ¹²⁴⁹

I.1.2. Monofluorophosphites

OCH₂CH₂OPF. (RO)₂PCl + b₁₈ 26°, ¹²⁰¹ d₄²⁰ 1.14039, MR_D 19.90, -124.4 ppm, J_{PF} 127

OCH(Me)CH₂OPF. (RO)₂PCl n_D²⁰ 1.4035, MR_D 24.

OCH(Me)CHMeOPF. (RO)₂PCl n_D²⁰ 1.4020, MR_D 29.

OCH(Me)CH₂CH₂OPF. (RO)₂PCl n_D²⁰ 1.4160, MR_D 29.

OCH₂.C(Et)(Bu)CH₂OPF. 1.1241, n_D²⁰ 1.4765, NMR. ¹²⁰¹

OCH₂(CH₂)₂CH₂OPF. (RO)₂PCl n_D²⁰ 1.4450, MR_D 30.

OCH₂(CH₂)₄CH₂OPF. (RO)₂PCl n_D²⁰ 1.4270, MR_D 39.

OCH₂(CH₂)₈CH₂OPF. (RO)₂PCl n_D²⁰ 1.4798, MR_D 57.

1,2-C₆H₁₀O₂PF. (RO)₂PCl n_D²⁰ 1.4586, MR_D 36.

1,2-C₆H₄O₂PF. (RO)₂PCl b₆ 36.5°, ¹²⁴⁹ b₆ 38, n_D²⁷ 1.5080, ¹²⁴⁹ ³¹P -119 F NMR, ¹²⁴⁹ 31P -1

JPOCCH ca. 1 Hz, J_P 3-Me-1,2-C₆H₃O₂PF. (RO)₂PCl n_D²⁰ 1.5170, MR_D 39.

4-Me-1,2-C₆H₃O₂PF. (RO)₂PCl n_D²⁰ 1.5220, MR_D 39.

1,2-C₆H₄C(:O)OPF. (RO)₂PCl n_D²⁵ 1.5390. ¹²⁴⁹

I.1.3. Dichlorophosphites

TYPE: ROPCl₂

CD₃OPCl₂. Ib. b₆₀ 31-MeOPCl₂. Ia. b₇₅₈ 95-1.47725, ²⁹⁹, ⁷⁴⁰ ³¹P

C₆H₅CH(CO₂Et)OPCl₂. Ia 1.2720, n_D²¹ 1.5259, (-)-mandelate α_D¹⁶ -

phosphorous Acid

List of Compounds 149

of RR'N). Named in connection with treatment of plastics.¹⁴⁴

Named in connection with plastic strips which

acidite. Used for the

acid. Catalyze the reaction to high molecular

as hexamethylene isocyanate. Improves octane

amidite. Is an additive for improving the octane number, deposits without lower-

ustriamide" "Di-Bu-N- as antiknock additives

with pigmented polyurethane, improving viscosity

2.628
b₁₈₀ 50°, n_D²⁶
0 ppm, J_{PF} 1295 Hz.¹²⁰⁹

n_D²⁰ 1.3400, ¹⁹F
287 Hz.^{1209, 1249}
42° ¹⁹F NMR, ¹²⁴⁹
09, ¹²⁴⁹
0 1.3580, ¹²⁴⁹ 31P

n_D²⁷ 1.4575, ¹⁹F
326 Hz.^{1209, 1249}
2 59°, n_D²³ 1.4488,
F 1328 Hz.^{1209, 1249}

I.1.2. Monofluorophosphites with P in Ring System

OCH₂CH₂OPF. (RO)₂PCl + SbF₃.^{1064, 1201, 1249} b₁₇₀ 48°, ¹²⁴⁹
b₁₈ 26°, ¹²⁰¹ d₄²⁰ 1.3552, ¹²⁰¹ n_D^{23.5} 1.4003, ¹²⁴⁹ n_D²⁰
1.4039, MR_D 19.90 (20.56), ^{1201, 1249} ¹⁹F NMR, ¹²⁴⁹ 31P
-124.4 ppm, J_{PF} 1223 Hz, ^{1209, 1249} ¹H NMR.⁴³²

OCH(Me)CH₂OPF. (RO)₂PCl + SbF₃. b₁₀₀ 44°, d₄²⁰ 1.2226,
n_D²⁰ 1.4035, MR_D 24.78 (25.18), ^{1064, 1201} IR.¹²⁰¹

OCH(Me)CHMeOPF. (RO)₂PCl + SbF₃. b₁₆ 28°, d₄²⁰ 1.1568,
n_D²⁰ 1.4020, MR_D 29.08 (29.79).¹²⁰¹

OCH(Me)CH₂CH₂OPF. (RO)₂PCl + SbF₃. b₁₆ 37°, d₄²⁰ 1.1857,
n_D²⁰ 1.4160, MR_D 29.22 (29.79).¹²⁰¹

OCH₂.C(Et)(Bu)CH₂OPF. (RO)₂PCl + SbF₃. b₁ 61°, d₄²⁰
1.1241, n_D²⁰ 1.4765, MR_D 52.28 (52.88), ¹²⁰¹ ¹H
NMR.¹²⁰¹

OCH₂(CH₂)₂CH₂OPF. (RO)₂PCl + SbF₃. b₁₆ 38°, d₄²⁰ 1.2180,
n_D²⁰ 1.4450, MR_D 30.16 (29.80), ¹²⁰¹ IR, ¹²⁰¹ ¹H NMR.¹²⁰¹

OCH₂(CH₂)₄CH₂OPF. (RO)₂PCl + SbF₃. b₁ 66°, d₄²⁰ 1.0840,
n_D²⁰ 1.4270, MR_D 39.94 (39.03).¹²⁰¹

OCH₂(CH₂)₈CH₂OPF. (RO)₂PCl + SbF₃. b₂ 80°, d₄²⁰ 1.1041,
n_D²⁰ 1.4798, MR_D 57.09 (57.50).¹²⁰¹

1,2-C₆H₁₀O₂PF. (RO)₂PCl + SbF₃. b₁ 34°, d₄²⁰ 1.2140,
n_D²⁰ 1.4586, MR_D 36.93 (36.83), ¹²⁰¹ ¹H NMR.¹²⁰¹

1,2-C₆H₄O₂PF. (RO)₂PCl + SbF₃.^{1201, 1249} or + NaF.¹²⁴⁹
b₆ 36.5°, ¹²⁴⁹ b₆ 38°, ¹²⁰¹ d₄ 1.3592, ¹²⁰¹ n_D²⁵ 1.5092,
n_D²⁷ 1.5080, ¹²⁴⁹ n_D²⁰ 1.5160, MR_D 35.13 (35.43), ¹²⁰¹
¹⁹F NMR, ¹²⁴⁹ 31P -123.1 ppm, J_{PF} 1305 Hz, ¹H NMR, ¹²⁰¹
J_{POCCH} ca. 1 Hz, J_{POCCCH} < 0.5 Hz.^{1209, 1249}

3-Me-1,2-C₆H₃O₂PF. (RO)₂PCl + SbF₃. b₂ 58°, d₄²⁰ 1.3045,
n_D²⁰ 1.5170, MR_D 39.94 (calc. 40.04).¹²⁰²

4-Me-1,2-C₆H₃O₂PF. (RO)₂PCl + SbF₃. b₇ 84°, d₄²⁰ 1.3150,
n_D²⁰ 1.5220, MR_D 39.92 (calc. 40.04).¹²⁰²

1,2-C₆H₄C(:O)OPF. (RO)₂PCl + KSO₂F. b_{0.15-0.2} 44-7°,
n_D²⁵ 1.5390.¹²⁴⁹

I.1.3. Dichlorophosphites

TYPE: ROPCl₂

CD₃OPCl₂. Ib. b₄₀ 31-2°, d₄²⁴ 1.3892, n_D²⁴ 1.4682.¹⁵⁷

MeOPCl₂. Ia. b_{7.53} 95-6°, d₄⁰ 1.4275, d₄²⁰ 1.3980, n_D⁰
1.47725, ^{199, 740} 31P -180.5, -181.0 ppm.^{396, 915}

C₆H₅CH(CO₂Et)OPCl₂. Ia. b₂ 105-8°, d₄⁰ 1.2827, d₄²¹
1.2720, n_D²¹ 1.5259, α_D⁸ -117.5° (l = 10 cm) from the
(-)-mandelate α_D⁶ -131.0°.⁴⁵⁰



07/714441

#3

[Handwritten signature]

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION TO REISSUE
U.S. PAT. NO. 4,912,155,
ISSUED MARCH 27, 1990

SERIAL NO.

FILED MAY _____, 1991

FOR ANTIOXIDANT AROMATIC FLUORO-
PHOSPHITES

"Express Mail" mailing label

number B 89910190

Date of Deposit JUNE 13, 1991

I hereby certify that this paper or fee is being
deposited with the United States Postal Service
"Express Mail Post Office to Addressee" service
under 37 CFR 1.10 on the date indicated above and
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Trademarks, Washington, D.C. 20231

DEBORAH REAGAN

(Typed or printed name of person mailing paper or
fee)

[Handwritten signature: Deborah Reagan]
(Signature of person mailing paper or fee)

OFFER TO SURRENDER PATENT UNDER 37 C.F.R. 1.178


Honorable Commissioner of Patent and Trademarks
Washington, D. C. 20231

Sir:

The undersigned representative of Ethyl Corporation which is now sole owner by assignment, and on whose behalf and with whose assent the accompanying application for the reissue of U.S. Patent No. 4,912,155, granted on March 27, 1990, is made, hereby offers to surrender said Patent when received. In this regard, attention is directed to the Letter, a photocopy of which is attached hereto, which assignee filed with the U.S. Patent and Trademark Office on or about June 8, 1990. Specifically, the U.S. Patent and Trademark Office has yet to provide the original patent. No response to the Letter has been received, although assignee has been verbally informed that the office is in the process of reprinting an original patent.

Ethyl Corporation

Date: May 20, 1991

By 
Roger A. Moser
Senior Vice President